Spin relaxation in quantum dots due to electron exchange with leads

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We calculate spin relaxation rates in lateral quantum dot systems due to electron exchange between dots and leads. Using rate equations, we develop a theoretical description of the experimentally observed electric current in the spin blockade regime of double quantum dots. Single expression fits the entire current profile and describes the structure of both the conduction peaks and of the suppressed ('valley') region. Extrinsic rates calculated here have to be taken into account for accurate extraction of intrinsic relaxation rates due to the spin-orbit and hyperfine spin scattering mechanisms from spin blockade measurements.

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During the last decade, considerable progress was made in development of devices utilizing spin degrees of freedom of electron systems,[1] promising manufacturing of functional devices. The ultimate usage of spin devices for quantum information processing[2] would capitalize on potentially weak coupling of spin systems with their environments. This coupling can be of two origins: (i) intrinsic to material; and (ii) device specific (geometric or extrinsic), due to interactions with leads, substrate, etc. Knowledge of spin relaxation and decoherence resulting from the coupling to environment is crucial for evaluating the limitations of the spin-based devices.

The intrinsic mechanisms responsible for spin relaxation in semiconductor devices are the hyperfine interaction with nuclear spins and spin-orbit interaction, for a review see Refs. 1, 3. Relaxation rates were experimentally analyzed for quantum dot devices on the basis of GaAs heterostructures. The resulting coherence and spin flip times can be as short as $10-100$ns in zero magnetic field, but are much longer than microseconds in the field[1]. For silicon dot structures such measurements have yet to be done; however the two intrinsic mechanisms are small in bulk Si, and these times are expected to be orders of magnitude longer, $\gtrsim$ 1second.[4, 5]

The goal of this paper is to analyze recent studies [6, 7, 8, 9] of charge transport through double quantum dot systems. Such systems are proposed for spin spectroscopy and control of dot states. The spin blockade, Fig. 1, of electric current through double quantum dots is one of the most striking signatures of the electronic spin degree of freedom in nanoelectronic devices. We examine in detail the electric current through a double quantum dot system. We point out that while recent studies of relaxation processes in double dots were focused on intrinsic mechanisms of spin relaxation, the current contains an essential contribution arising from the extrinsic mechanism of spin relaxation as a consequence of electron tunneling between dots and leads. Below we propose a theoretical description of this effect.

We show that the current remains finite, and equal to $I = e\Gamma_{\rm s}/3$, even in the absence of intrinsic spin relaxation mechanisms. Parameter $\Gamma_{\rm s}$ is the *extrinsic* spin relaxation rate due to electron tunneling to the leads. This rate strongly depends on the relative energy between the spin state in the dot, $E = E_{\uparrow,\downarrow}$, and the Fermi level, E_F, of the connected lead

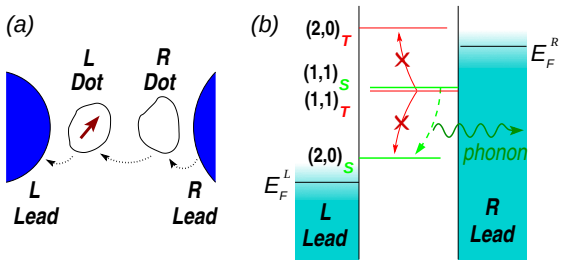

FIG. 1: (Color online) (a) Lateral double dot system in the spin blockaded regime. In the initial configuration, the system has one electron on the left dot, $(1,0)$. Electric current through the dot flows from right to left in a sequence of steps. (b) Energy level representation of transport through the dot. The sequence of transitions via singlet states, $(1,0) \to (1,1)_{\rm S} \rightsquigarrow (2,0)_{\rm S} \to (1,0)$, quickly transports electron from the right to left lead, whereas current through intermediate triplet state, $(1,0) \to (1,1)_{\rm T}$, is blocked since subsequent transition to $(2,0)_{\rm S(T)}$ state is forbidden by spin (energy) conservation.

$$\Gamma_{\rm s} = \Gamma_{\rm R}[1 - f(\Delta)] + 2\gamma_2(\Delta)\,, \qquad (1)$$
$$\Delta = E - E_F\,, \quad \gamma_2 = \Gamma_{\rm R}^2 \frac{T}{\Delta^2}\frac{\hbar}{2\pi}\,,$$

where $f(\Delta)$ is Fermi function of electrons in the lead and γ_2 is written for large separation from the Fermi level. The first term dominates when $\Delta \gtrsim -T$ (T is the temperature) and describes the probability of emptying a given spin state back into the lead, after which the loading from the lead can start again. This term is linear in the tunnelling rate $\Gamma_{\rm R}$ between the quantum dot and its lead. The second contribution γ_2 to the spin flip transition rate in Eq. (1), commonly referred to as co-tunnelling, describes the mechanism similar to the Korringa relaxation mechanism[10] of localized magnetic moments in bulk conductors due to their coupling to the spins of itin-

erant electrons. Note that although this term contains two additional small parameters, $T/|\Delta|$ and $\Gamma_{\rm R}/|\Delta|$, for a deep level, *i.e.* $\Delta \ll -T$, it vanishes only as $1/|\Delta|^2$, compared to the exponential decay, $\exp(-|\Delta|/T)$, of the first term. We use Eq. (1) to analyze experimental data of Ref.[8] and obtain an almost perfect agreement between the measured current and the current exclusively due to extrinsic relaxation processes.

Previously, the influence of leads on processes in dots has been addressed in connection to different phenomena, such as the Kondo spin relaxation [11], cotunneling [12] transport and spin relaxation in a single dot [13, 14, 15, 16, 17] and nuclear spin relaxation in the Coulomb blockade regime [18].

The spin blockade occurs because transport through a double dot, Fig. 1(a), can only flow via singlet states and is blocked if electron entering the right dot from the right lead forms a triplet with the spin on the left dot.[1, 19] The relevant energy states corresponding to this regime are shown in Fig. 1(b), where $(n_L, n_R)_{\rm S,T}$ denotes a two-electron state with $n_{L(R)}$ electrons on the left (right) dot in singlet (S) or triplet (T) configuration. States $(2,0)_{\rm S,T}$ are separated by level spacing in the left dot, whereas smallness of interdot exchange leaves states $(1,1)_{\rm S,T}$ nearly degenerate. Relaxation of the spin lifts the spin blockade.[20, 21, 22] After time $\tau_{\rm s}$ triplet state $(1,1)_{\rm T}$ can relax by spin flip into singlet $(1,1)_{\rm S}$, and thus the current $I \sim e/\tau_{\rm s}$ can be used to experimentally determine the lifetime of an electron spin in a device, *c.f.* [23]. The importance of the spin relaxation due to electron exchange with the leads is indicated by significant currents on the boundaries of the spin blockade region that has been observed in both GaAs[7] and silicon[8, 9] lateral double dots. Moreover, this mechanism may dominate spin relaxation in silicon quantum dots, where intrinsic mechanisms are weak [3, 4, 5].

The full Hamiltonian for the double dot, $\mathcal{H} = H_{\rm d} + H_{\rm l} + V$, consists of interacting electrons in the dots, $H_{\rm d}$, free electrons in the leads, $H_{\rm l}$, and the tunnelling between the leads and the dots, V. Here we do not specify the exact form of the Hamiltonian $H_{\rm d}$ for strongly interacting electron states in the double dot system, which in principle can be written in terms of the creation, $d^\dagger_{\alpha\,\sigma}$, and annihilation, $d_{\alpha\,\sigma}$, operators in the left and right dots ($\alpha = L, R$). We only assume that the lowest eigenstates of $H_{\rm d}$ have the following hierarchy of energies $E_{(2,0)_{\rm S}} < E_{(1,1)} < E_{(2,0)_{\rm T}}$, as illustrated in Fig. 1(b). The Hamiltonian of free electrons in the leads $H_{\rm l}$ is written in terms of the creation, $c^\dagger_{\alpha\,\mathbf{k}\,\sigma}$, and annihilation, $c_{\alpha\,\mathbf{k}\,\sigma}$, operators of electrons in lead α with momentum $\mathbf{k}$, spin σ and energy $\xi_{\alpha\,\mathbf{k}}$: $H_{\rm l} = \sum_{\alpha=L,R}\sum_{\mathbf{k},\sigma} \xi_{\alpha\,\mathbf{k}} c^\dagger_{\alpha\,\mathbf{k}\,\sigma} c_{\alpha\,\mathbf{k}\,\sigma}$. The coupling between states in lead α to electron states in the dot is represented by the tunnelling Hamiltonian $\hat{V}$, with the tunnelling probabilities $W_{\alpha\,k}$:

$$V = \sum_{\alpha=L,R}\sum_{\mathbf{k},\sigma} \left(W_{\alpha\,k} d^\dagger_{\alpha\,\sigma} c_{\alpha\,\mathbf{k}\,\sigma} + W^*_{\alpha\,k} c^\dagger_{\alpha\,\mathbf{k}\,\sigma} d_{\alpha\,\sigma} \right) . \quad (2)$$

The relevant dot states that are involved in electron current are single-particle states with an electron on the left dot, $|\uparrow 0\rangle = d^\dagger_{L\uparrow}|0\rangle$, $|\downarrow 0\rangle = d^\dagger_{L\downarrow}|0\rangle$, and two-particle states with one electron on each dot, that can be in either singlet, $|S\rangle = \frac{1}{\sqrt{2}}(d^\dagger_{R\uparrow} d^\dagger_{L\downarrow} - d^\dagger_{R\downarrow} d^\dagger_{L\uparrow})|0\rangle$, or one of the triplet spin configurations, $|T_0\rangle = \frac{1}{\sqrt{2}}(d^\dagger_{R\uparrow} d^\dagger_{L\downarrow} + d^\dagger_{R\downarrow} d^\dagger_{L\uparrow})|0\rangle$, $|T_+\rangle = d^\dagger_{R\uparrow} d^\dagger_{L\uparrow}|0\rangle$, $|T_-\rangle = d^\dagger_{R\downarrow} d^\dagger_{L\downarrow}|0\rangle$. The system can be in one of these states and the corresponding probabilities satisfy the normalization: $P_{\uparrow 0} + P_{\downarrow 0} + P_S + P_{T_0} + P_{T_+} + P_{T_-} = 1$. We omit the $(2,0)$ states, since $P_{(2,0)} \approx 0$ due to fast escape to left lead. The rate equations for two-particle states in the dot are

$$\dot{P}_S = -(\gamma_1 + \frac{3}{2}\gamma_2)P_S + \frac{1}{2}\bar{\gamma}_1(P_{\uparrow 0} + P_{\downarrow 0}) + \frac{1}{2}\gamma_2(P_{T_0} + P_{T_+} + P_{T_-}) - \Gamma P_S\,, \quad (3a)$$

$$\dot{P}_{T_0} = -(\gamma_1 + \frac{3}{2}\gamma_2)P_{T_0} + \frac{1}{2}\bar{\gamma}_1(P_{\uparrow 0} + P_{\downarrow 0}) + \frac{1}{2}\gamma_2(P_S + P_{T_+} + P_{T_-})\,, \quad (3b)$$

$$\dot{P}_{T_+} = -(\gamma_1 + \gamma_2)P_{T_+} + \bar{\gamma}_1 P_{\uparrow 0} + \frac{1}{2}\gamma_2(P_S + P_{T_0})\,, \quad (3c)$$

$$\dot{P}_{T_-} = -(\gamma_1 + \gamma_2)P_{T_-} + \bar{\gamma}_1 P_{\downarrow 0} + \frac{1}{2}\gamma_2(P_S + P_{T_0})\,. \quad (3d)$$

For instance, Eq. (3a) describes the change of the singlet state population. P_S is reduced by the transitions to states $|\uparrow 0\rangle, |\downarrow 0\rangle$ (with rates $\gamma_1/2$), but is increased by the reverse transitions (with rates $\bar{\gamma}_1/2$). It also couples to all triplet states, $|T_0\rangle$ and $|T_\pm\rangle$, with a single rate, $\gamma_2/2$. Additionally, we include the possibility of transition from singlet $|S\rangle = (1,1)_S$ to $(2,0)_{\rm S}$ between the two dots, with rate Γ, by introducing $-\Gamma P_S$ term. The remaining equations in system (3) for triplet states have similar structure, but without the interdot transitions.

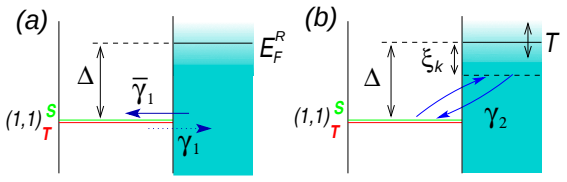

FIG. 2: (Color online) Electron exchange with the lead in lowest tunnelling orders. (a) In the first order process, a single electron jumps on or off the dot, with electron energy in the lead equal to $E_{(1,1)}$. The de-tuning from the Fermi level is $\Delta = E_{(1,1)} - E_F^R$. (b) The second order process represents double electron tunnelling through a virtual state. Energy of participating electrons in the lead are close to the lead Fermi energy and can be far from the dot energy level.

The transition rates in Eqs.(3) are given by the lowest two orders in tunnelling, Fig. 2, and describe (a) electron hopping on ($\bar{\gamma}_1$) and off (γ_1) the dot

$$\bar{\gamma}_1 = \Gamma_{\rm R} f(\Delta)\,, \quad \gamma_1 = \Gamma_{\rm R}\,[1 - f(\Delta)]\,; \tag{4}$$

and (b) double exchange of electrons between the lead and the dot (γ_2), via virtual states $|\uparrow 0\rangle, |\downarrow 0\rangle$, resulting in the creation of an electron-hole pair in the lead,

$$\gamma_2 = \frac{2\pi}{\hbar}\sum_{\mathbf{k}} \mathcal{N}_F |W_{R,k}|^4 \left|\frac{1}{\Delta - \xi_k + i0}\right|^2 f(\xi_k)[1 - f(\xi_k)]\,.$$

Here $f(\xi) = 1/(1+\exp(\xi/T))$ is the Fermi function in the right lead, and $\Gamma_{\rm R} = (2\pi/\hbar)\mathcal{N}_F\,|W_{R,k}|^2$, where $\mathcal{N}_F$ is the density of states in the lead near the Fermi level ($\Delta, \xi \ll E_F$). The expression for γ_2 formally diverges at $\xi_k = \Delta$, but is applicable for $|\Delta|$ large compared with temperature, when the contribution from $\xi_k \approx \Delta$ is exponentially suppressed [24]:

$$\gamma_2 \approx \Gamma_{\rm R}\frac{T\,T_{\rm R}}{\Delta^2}, \quad T_{\rm R} = \frac{\hbar\Gamma_{\rm R}}{2\pi}, \quad |\Delta| \gtrsim T. \tag{5}$$

To conclude discussion of Eqs. (3)-(5), we note that they were obtained from general transition rate equations for diagonal elements of the full density matrix. Each such element corresponds to an eigenstate of the full lead-dot system, e.g. $|\,i\,\rangle = |\,e_i\,\rangle \times |\,{\rm dot}_i\,\rangle$ with energy ϵ_i. Transition rates between these states are,

$$\Gamma^{fi} = \frac{2\pi}{\hbar}\delta(\epsilon_f - \epsilon_i)\left|V_{fi} + \sum_m \frac{V_{fm}V_{mi}}{\epsilon_i - \epsilon_m + i0} + \dots\right|^2 .$$

Since the environment relaxes much faster than the dot we take the trace over the electronic configurations $\{e_i, e_f\}$ in the leads, and define $\gamma_{fi} = {\rm Tr}_{e_i,e_f}\,\Gamma^{fi}\,\rho^0_{e_f}\rho^0_{e_i}$ as the rate of transition between the *dot* states $|\,{\rm dot}_i\,\rangle$ and $|\,{\rm dot}_f\,\rangle$. Here ρ^0_e is the equilibrium density matrix for non-interacting electrons in the leads.

We assume that the lead-dot tunnelling rates $\Gamma_{\rm R,L}$ are larger than the interdot rate Γ. Indeed, the tunnelling between the dots is accompanied by the emission of a phonon (or another excitation) that carries away the energy difference between $E_{(1,1)_{\rm S}}$ and $E_{(2,0)_{\rm S}}$. Such coupling of electron states to phonon modes results in additional smallness of the rate Γ, which is determined by both the overlap between electron states in the two dots and the matrix elements of electron-phonon coupling. However, a microscopic derivation of Γ is beyond the scope of this paper.

In the absence of magnetic fields, states that differ only by spin projections, are degenerate, and we introduce $P_0 = P_{\uparrow 0} = P_{\downarrow 0}$ and $P_{T_1} = P_{T_+} = P_{T_-}$. We use the normalization to remove redundant P_0, and obtain a system of equations for only three variables, P_{T_0}, P_{T_1} and P_S.

In the limit of negligible tunnelling to the left dot ($\Gamma \to 0$), this system of equations can be diagonalized:

$$\dot{P}_\eta(t) + \Gamma_\eta P_\eta(t) = J_\eta\,, \quad \eta = 1,2,3: \tag{6}$$
$$\begin{array}{llll} P_1 = P_{T_0} - P_{T_1}, & \Gamma_1 = \Gamma_{\rm s}, & J_1 = 0; \\ P_2 = 3P_S - (P_{T_0} + 2P_{T_1}), & \Gamma_2 = \Gamma_{\rm s}, & J_2 = 0; \\ P_3 = P_S + (P_{T_0} + 2P_{T_1}), & \Gamma_3 = \Gamma_{\rm c}, & J_3 = 2\bar{\gamma}_1. \end{array}$$

The first two eigenmodes describe dynamics of spin in the double dot system, with the spin flip rate $\Gamma_{\rm s}$. The last eigenmode is for the total occupation of the right dot by one electron, with characteristic charge relaxation rate $\Gamma_{\rm c}$. The two rates are,

$$\Gamma_{\rm s} = \gamma_1 + 2\gamma_2\,, \quad \Gamma_{\rm c} = \Gamma_{\rm R}[1 + f(\Delta)]. \tag{7}$$

Here we remark on spin relaxation in a single dot. The relevant states are empty (P_0) and singly occupied with spin up/down ($P_{\uparrow/\downarrow}$). One finds that Eqs. (3) for these states are modified, but the dynamics of $(P_\uparrow - P_\downarrow)$ and $(P_\uparrow + P_\downarrow)$ is given by Eq. (6) with $\eta = 1$ and $\eta = 3$ respectively, with unchanged $\Gamma_{\rm s}, \Gamma_{\rm c}$ and the source terms.

The current through the right dot is defined as the rate of charge escape from the singlet state into the left dot, $I(t) = e\Gamma P_S(t)$, and the stationary solution of rate equations, Eqs. (3), gives the steady current

$$I = e\frac{f(\Delta)}{1+f(\Delta)}\frac{2\Gamma\Gamma_{\rm s}}{4\Gamma_{\rm s} + \Gamma(3 + \Gamma_{\rm s}/\Gamma_{\rm c})}\,. \tag{8}$$

This equation describes the magnitude of current through a lateral double quantum dot in the regime of spin blockade. We plot typical current profiles in Fig. 3 as a function of de-tuning Δ. In experiments Δ is controlled by gate voltages and current I is mapped as a function of these voltages[7, 8].

Equation (8) has particularly simple form in two important limits,

$$\Gamma_{\rm s} \gg \Gamma\ : \quad I = \frac{1}{2}e\Gamma\frac{f(\Delta)}{1+f(\Delta)}, \quad ({\rm peak})\,, \tag{9a}$$

$$\Gamma_{\rm s} \ll \Gamma\ : \quad I = \frac{1}{3}e\Gamma_{\rm s}, \quad ({\rm valley},\ f(\Delta) \approx 1)\,. \tag{9b}$$

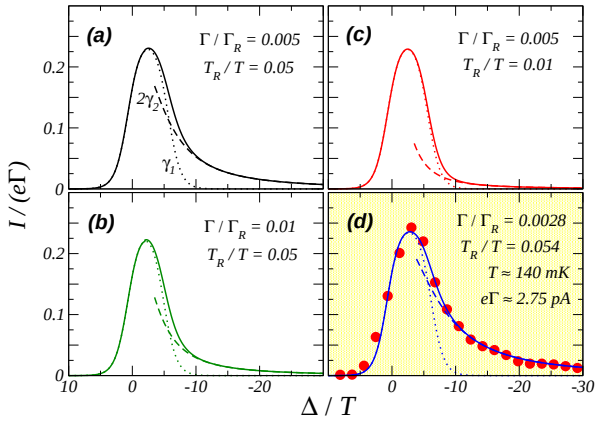

FIG. 3: (Color online) Current through a double dot system in the spin blockaded regime. The dotted (dashed) lines show asymptotes due to first (second) order processes that dominate peak(valley) and are given by Eq. (8) with Γ_s replaced by γ_1 ($2\gamma_2$). Panel (d) shows a fit to measured current (circles) along a line-cut of the spin blockade peak reported in [8].

The first limit describes the left slope and the peak of the current, Fig. 3. As a consequence, the dimensionless current $I/e\Gamma$ is nearly independent of system parameters. Its meaning is straightforward: the right dot is loaded by one electron with probability $2f(\Delta)/(1+f(\Delta))$, as follows from Eqs.(6) and (7), and the singlet state is 1/4 of this probability, as all two-particle states are equally populated when singlet-triplet relaxation is fast. The current is proportional to the escape rate Γ.

The second limit, Eq. (9b), describes the spin blockade region. Depending on parameters it can exhibit distinct non-exponentially decaying tail for $|\Delta|/T \gg 1$, see Fig. 3, that should be easily observable. The factor 1/3 comes from the probability of the system to be in one of the triplet states. Indeed, according to the stationary solution of Eqs.(3) in the limit $|\Delta| \gg T$, the right dot is definitely occupied. The probabilities of finding the system in the singlet and triplet states are determined by the ratio of Γ and Γ_s,

$$\begin{pmatrix} P_S \\ P_{T_0} \\ P_{T_1} \end{pmatrix} = \frac{1}{4\Gamma_s + 3\Gamma} \begin{pmatrix} \Gamma_s \\ \Gamma_s + \Gamma \\ \Gamma_s + \Gamma \end{pmatrix} .$$

For $\Gamma_s \gg \Gamma$ the equilibrium is $P_S = P_{T_{0,1}} = 1/4$, as we discussed. On the other hand, if $\Gamma \gg \Gamma_s$ the singlet state is almost empty ($P_S \sim \Gamma_s/3\Gamma$) since it takes time $1/\Gamma_s$ to populate this state from one of the triplet states while it quickly empties into the left dot. The triplet states are all equally populated, each with probability $\approx 1/3$.

Finally, we note that the second conduction peak observed in [7, 8] is explained by complementary hole transport in the cycle $(2,1) \to (1,1)_{(T\to)S} \to (2,0)_S \to (2,1)$. In this cycle, the spin flip relaxation between $(1,1)_{T,S}$ states occurs due to electron exchange between the left dot and the left lead via intermediate states $(2,1)$.

In conclusion, we presented a model for spin relaxation due to electron exchange between dots and leads. We used it to construct a theory of current through a spin blockaded double quantum dot, where spin flips result in transitions between $(1,1)_S$ and $(1,1)_T$ states, with rate $\Gamma_s = \gamma_1 + 2\gamma_2$. To estimate the resulting relaxation times, we neglect intrinsic relaxation rate γ_{intr} (which in principle can be incorporated in Eq. (8) by replacement $\gamma_2 \to \gamma_2 + \gamma_{intr}$), and use this theory to extract relevant times from experiment on silicon double dots[8]. We obtain $\tau_s \sim 0.1$ ns in the peak of the current and $\tau_s \sim 1\,\mu$s in the valley, see Fig. 3(d). These relaxation times are comparable and even shorter than those due to intrinsic spin-orbit and hyperfine mechanisms. The coupling between electron states in the dots and the leads has to be taken into account for analysis of intrinsic spin relaxation mechanisms from the spin blockade measurements.

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[24] The estimate in Eq. (5) becomes invalid for $|\Delta| \lesssim T$. However the total spin relaxation rate is a sum of γ_1 and γ_2, see Eq. (7) later, and γ_1 dominates the relaxation in the range $\Delta \gtrsim -T\ln(T/T_{\rm R})$ with $T \gg T_{\rm R}$, so γ_2 is to be omitted there anyway. We write the rate equations assuming that these two rates coexist in entire range of Δ. In the end, however, the contribution of γ_2 should be taken into account when Δ satisfies the applicability condition of Eq. (5).